

## Aerosol optical properties during INDOEX based on measured aerosol particle size and composition

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[1] The light scattering and light absorption as a function of wavelength and relative humidity due to aerosols measured at the Kaashidhoo Climate Observatory in the Republic of the Maldives during the INDOEX field campaign has been calculated. Using size-segregated measurements of aerosol chemical composition, calculated light scattering and absorption has been evaluated against measurements of light scattering and absorption. Light scattering coefficients are predicted to within a few percent over relative humidities of 20–90%. Single scattering albedos calculated from the measured elemental carbon size distributions and concentrations in conjunction with other aerosol species have a relative error of 4.0% when compared to measured values. The single scattering albedo for the aerosols measured during INDOEX is both predicted and observed to be about 0.86 at an ambient relative humidity of 80%. These results demonstrate that the light scattering, light absorption, and hence climate forcing due to aerosols over the Indian Ocean are consistent with the chemical and physical properties of the aerosol at that location. *INDEX TERMS*: 0305 Atmospheric Composition and Structure: Aerosols and particles (0345, 4801); 0345 Atmospheric Composition and Structure: Pollution—urban and regional (0305); 0360 Atmospheric Composition and Structure: Transmission and scattering of radiation; *KEYWORDS*: aerosol, INDOEX, single-scattering albedo, extinction coefficient

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### 1. Introduction

[2] Airborne particles are important to an understanding of both the direct and possible indirect effects of mankind's activities on climate (climate forcing). Direct effects of atmospheric particles include the scattering of sunlight back to space and therefore a decrease in the amount of light reaching the ground. In addition, light absorption by airborne particles converts light into thermal energy, which leads to heating of the atmosphere while reducing the light that reaches the land or ocean surface. Changes in the concentration of cloud condensation nuclei may modify the size and properties of cloud droplets [Rosenfeld *et al.*, 2001]. Both of these latter effects could be expected to indirectly affect climate by changing the evaporation and precipitation of water.

[3] The potential for aerosol forcing of climate is expected to vary according to regional differences in air

pollutant concentration and chemical composition. Areas with high predicted climate forcing by sulfate aerosols, for example, include the northeastern United States, Eastern Europe, and China among others. Much less is known about aerosol concentrations and chemical compositions in the area of the Indian Subcontinent and the Indian Ocean, but common experience and reports from the area suggest that air pollution levels at times can be severe. In order to more quantitatively define the effect of airborne particles on light scattering and absorption over the Indian Ocean, the INDOEX experiments were conducted during the period of the late 1990s [Ramanathan *et al.*, 1996, 2001]. The purpose of the present paper is to report the results of closure calculations. A detailed model calculation of aerosol light scattering and absorption is driven by the measurements of the size distribution and chemical composition of airborne particles sampled using cascade impactors over the period of 11–26 February 1999 at Kaashidhoo Island in the Republic of the Maldives. Comparisons are made to direct measurements of light scattering, light absorption, single scattering albedo, and growth in scattering as a function of relative humidity. Models for both an internally mixed and externally mixed aerosol are considered, and the relative importance of different chemical species to the atmospheric light extinction coefficient is presented for the externally mixed aerosol case. This analysis employs the only data in existence on the size distribution of black light-absorbing elemental carbon particles in this part of the world and thus is of particular

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interest to explaining the high levels of light absorption in the atmosphere near the surface of the ocean downwind of the Indian Subcontinent.

## 2. INDOEX Field Measurements

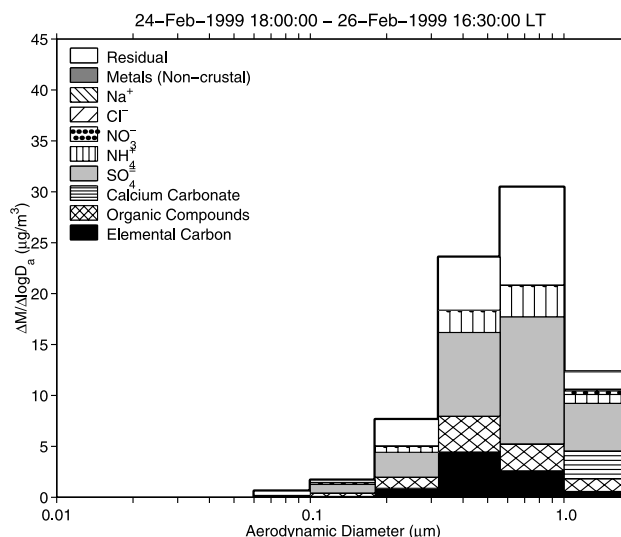
[4] Aerosol measurements were conducted at the Kaashidhoo Climate Observatory (KCO) on Kaashidhoo Island in the Republic of the Maldives in February of 1999 as part of the INDOEX field experiment [Ramanathan *et al.*, 2001; Chowdhury *et al.*, 2001; Lobert and Harris, 2002; Ramanathan *et al.*, 1996]. Particulate matter samples were collected with filter samplers and cascade impactors over consecutive 2-day sampling periods between 11 and 26 February 1999. Micro-orifice (MOUDI) impactors were used to collect particle samples in six size ranges that span the interval from 1.8  $\mu\text{m}$  to 0.056  $\mu\text{m}$  particle aerodynamic diameter ( $D_a$ ). There were four impactors at the sampling site, two with Teflon impactor substrates and two with aluminum foil impactor substrates. Measurements of fine particle ( $D_a \leq 1.8 \mu\text{m}$ ) concentration and total particle concentration (no size discrimination) were made with filter samplers. Complete details of the sampling and chemical characterization of the aerosol including a description of the uncertainties in the measurements are given by Chowdhury *et al.* [2001]. Figure 1 illustrates one example of the detailed data on aerosol size and chemical composition that is available for each sampling period.

[5] The impactor samples collected on Teflon substrates were analyzed to determine the aerosol mass, trace element, and ionic species concentrations. Gravimetric techniques were used to determine the aerosol mass concentration at 40% RH, ion chromatography was used for the analysis of nitrate, sulfate, and chloride, and ammonium ion concentrations were determined by colorimetry. From the impactor samples collected on aluminum foil substrates, organic and elemental carbon concentrations were determined by the thermo-optical technique of Birch and Cary [1996] as modified for impactor samples by Kleeman *et al.* [1999]. Organic compound mass was then estimated as 1.4 times the mass of organic carbon present on each impactor stage. The chemical analysis of the impactor samples provides chemical composition data for each size fraction collected on each separate impactor stage.

[6] To describe the aerosol size distribution, the gravimetric mass concentration data from the impactors were used and the aerodynamic particle diameters ( $d_a$ ) of the impactor stages were converted to physical particle diameters ( $d_p$ ). The conversion was made with the following relationship using the mean particle density at 70% relative humidity, which was determined from the chemical composition to be  $1.55 \text{ g cm}^{-3}$ .

$$d_p = d_a / \sqrt{\rho} \quad (1)$$

Due to the strong dependence of Mie scattering efficiencies on diameter, each impactor stage was subdivided into a set of five uniformly spaced  $\log(\text{diameter})$  intervals, and one fifth of the aerosol volume distribution was assigned to each of these narrower intervals. The particle mass distribution was converted to a particle number distribution using the particle radius at the center of each narrow diameter interval



**Figure 1.** Aerosol size and chemical composition as measured with a cascade impactor at Kaashidhoo Island on 24 February 1999.

and particle density as calculated from the aerosol chemical composition at each particle size.

[7] The impactor samples provide a detailed account of aerosol properties for particles less than 1.8  $\mu\text{m}$  in aerodynamic diameter. Particles larger than this may contribute to light extinction, and for limited comparisons against nephelometer measurements that include particles up to 10  $\mu\text{m}$  in diameter (PM10), larger particles must be included in the light extinction calculation. Filter samples that were collected along side the impactor samples provide measurements of the aerosol mass concentration for particles less than 1.8  $\mu\text{m}$  in diameter and for the total suspended particulate matter (TSP). The mass concentration of particles larger than 1.8  $\mu\text{m}$  aerodynamic diameter can be calculated by the difference between these two sets of filter samples. It is reasonable to assume that much of the particle mass in sizes greater than 1.8  $\mu\text{m}$  diameter consists of suspended dust [d'Almeida and Schutz, 1983]. Therefore the aerosol size distribution of suspended dust, as reported by Christoforou *et al.* [1996] was used to shape the measured aerosol mass for particle sizes larger than 1.8  $\mu\text{m}$  aerodynamic diameter. This coarse particle size distribution was then used to extend light scattering calculations to include particles up to 10  $\mu\text{m}$  aerodynamic diameter whenever comparisons were made against a nephelometer that viewed a PM10 aerosol sample.

[8] Aerosol light scattering coefficients were measured with two integrating nephelometers (model 3563, TSI, Inc., St. Paul, MN, USA) in series. A diffusion drier and heater upstream of the first nephelometer enabled a reference scattering measurement at a relative humidity of about 40%. Air leaving the first nephelometer passed through a humidifier that allowed the relative humidity in the second nephelometer to be scanned over relative humidities of up to 90% on an hourly basis. Two inertial impactors immediately downstream of the heater, together with a ball valve, enabled measurements on two particle size ranges (aerodynamic diameters less than 10  $\mu\text{m}$  and 1  $\mu\text{m}$ ) during

each hour. Further details of this “humidograph” system are given by *Carrico et al.* [1998] and *Anderson et al.* [1996].

[9] A continuous light absorption photometer (model PSAP, Radiance Research, Seattle, WA, USA) was used to determine the light absorption coefficient of the dehumidified particles in the two size ranges. The PSAP operates at a wavelength of 565 nm, and the nephelometers operated at wavelengths of 450, 550, and 700 nm. The PSAP and nephelometer data were corrected for instrument nonidealities according to the procedures described by *Bond et al.* [1999] and *Anderson and Ogren* [1998], respectively.

### 3. Model Description

[10] The light scattering and absorption calculations rely on Mie theory, which requires knowledge of the refractive index and aerosol size distribution. As described in the previous section, the aerosol size distribution is obtained by converting cascade impactor data on particle mass and chemical composition as a function of size into an equivalent population of particles having those same properties. Light extinction calculations were performed for what we will call internally and externally mixed particle populations with the true atmospheric aerosol existing somewhere between. In the model for an internally mixed aerosol, it is assumed that all particles of a particular size have the same chemical composition as the average composition measured from cascade impactor samples taken of particles of that size. These are modeled as well mixed, and another set of calculations for an internally mixed shell and core used a core of the insoluble materials with a shell of the soluble material. In the model for an externally mixed aerosol, it is assumed that each particle contains only a single chemical substance such as ammonium sulfate or organic compounds. The complex mixture of chemical substances present in particles of a given size is then represented as an external mixture of the appropriate number of distinctly different particle types. The method of calculating refractive indices and light scattering that is described in this paper has been discussed previously by *Larson et al.* [1988] and *Eldering et al.* [1994, 1993]. *Eldering et al.* [1993] demonstrated that skylight radiances calculated by these methods compare well to those measured independently with a radiometer.

[11] The total atmospheric light extinction coefficient can be expressed as the sum of four contributions:

$$\sigma_{ext} = \sigma_{sp} + \sigma_{ap} + \sigma_{sg} + \sigma_{ag}. \quad (2)$$

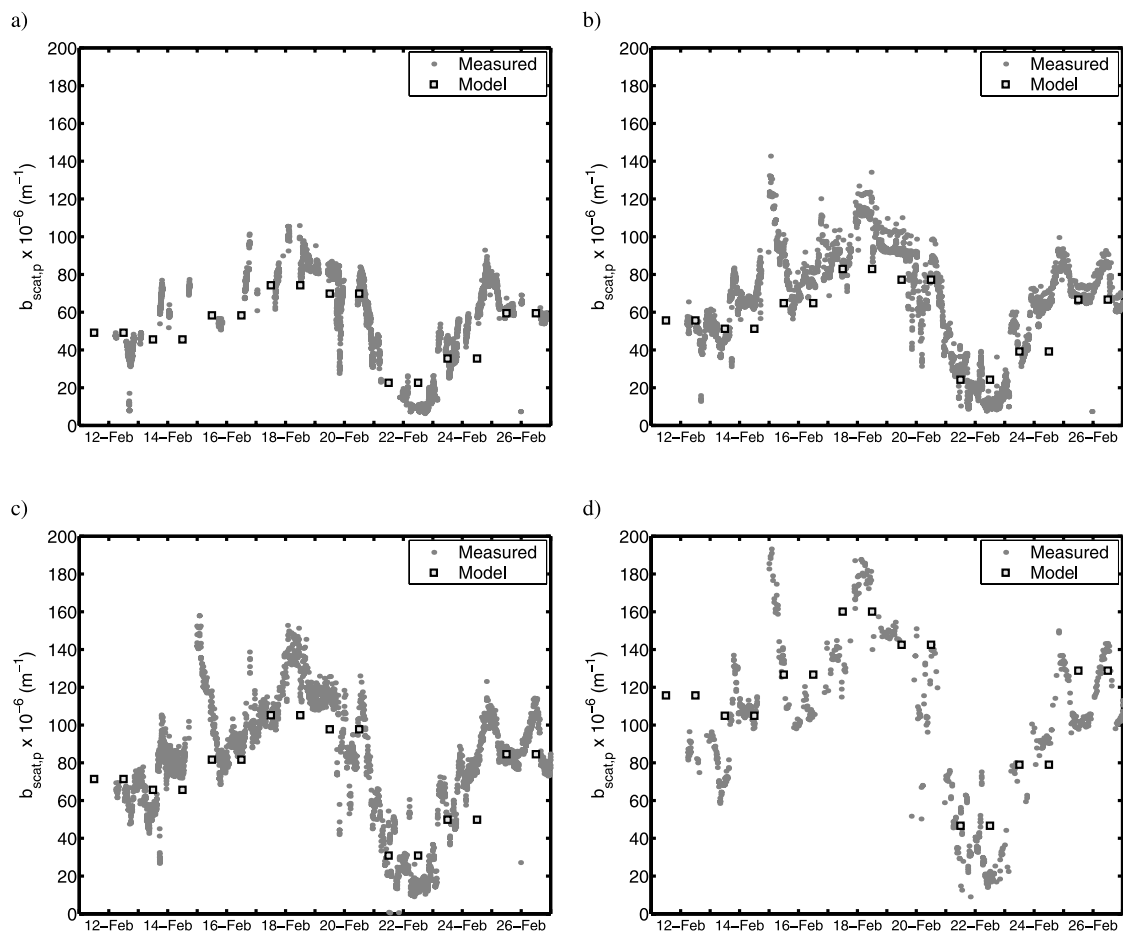
The extinction coefficient,  $\sigma_{ext}$ , is the sum of components due to light scattering by particles ( $\sigma_{sp}$ ), light absorption by particles ( $\sigma_{ap}$ ), scattering of light by gases (Rayleigh scattering,  $\sigma_{sg}$ ), and light absorption by gases ( $\sigma_{ag}$ ). Formulation of the Mie equations to determine scattering and absorption efficiencies for a given particle size and refractive index can be found in references such as *Van de Hulst* [1981] and *Bohren and Huffman* [1983]. Light scattering and absorption by particles are calculated from Mie theory, which requires real and imaginary refractive indexes and aerosol size distributions as described in more detail below. These Mie calculations also assume that the

particles are uniform spheres, a reasonable assumption for particles with significant water content.

[12] Estimation of the aerosol liquid water content as a function of particle size, composition, and RH is obtained using the relative humidity dependence model reported by *Sloane* [1984]. The concentration of soluble aerosol mass in each particle is calculated from the chemical composition as measured with impactors or filter samplers. The Köhler equation is applied using a semiempirical formulation of the equation for the activity of water that makes use of a van't Hoff factor for “typical” aerosols reported by *Sloane* [1984, Table 2]. As a result, the growth of each particle's diameter can be calculated as a function of relative humidity and the water soluble fraction of the particle.

[13] Estimation of the refractive index of each particle is required for the Mie calculation. For the externally mixed aerosol model, where each particle is composed of only one substance, the refractive index of each specific chemical substance is used in the Mie calculation. For water soluble species, the volume-weighted average refractive index of the single species and the added water is used. In the model for an internally mixed aerosol, the refractive index of each particle is calculated according to the volume-weighted average of the refractive indices of the components that are mixed within the particle. The chemical composition, species densities, and refractive index value of individual chemical compounds [*Weast*, 1985; *Sloane*, 1984] as tabulated by *Larson et al.* [1988] are combined to find the weighted average of the refractive indices. The weighting factors are the volume fraction contributed by each chemical substance. As the relative humidity increases and a particle grows, water is added to the mixture within the particle and the volume fraction of each individual species is recalculated along with the refractive index. In the shell and core calculations, the shell is assigned the volume average refractive index of the soluble materials and the core is assigned the volume average refractive index of the insoluble materials.

[14] In field experiments, aerosol size distributions may be measured by aerodynamic separation (impactors), optical techniques (optical particle counters), or electrical mobility (electrical aerosol analyzers or differential mobility analyzers). Knowledge of the aerosol size distribution is required for the Mie scattering calculation. The scattering and absorption efficiencies are calculated for each defined diameter interval of the aerosol particle size distribution according to the detailed implementation of the Mie equations as developed and described by *Toon and Ackermann* [1981]. In the internally mixed case,  $\sigma_{sp}$  and  $\sigma_{ap}$  are calculated by integrating the product of the scattering and absorption efficiency factors for each particle size and the number of particles per unit volume of air at a given size over the diameter range spanned by the particle size distribution. In the externally mixed case,  $\sigma_{sp}$  and  $\sigma_{ap}$  are calculated by integrating the product of the scattering and absorption efficiency factors for particles of a specific size composed of a single substance over the number of particles of that size and type per unit volume of air over the diameter range spanned by particles of that type. These light scattering and absorption coefficient increments due to particles of a particular chemical substance are then summed over all of the different particle types present in the external mixture to



**Figure 2.** Light scattering coefficients at 550 nm due to particles less than 1  $\mu\text{m}$  in diameter measured with a nephelometer and modeled from the size distributed impactor chemistry. The samples were collected in 1999 at Kaashidhoo Island. Panel (a) is at 30% RH, panel (b) at 50% RH, panel (c) at 70% RH, and panel (d) at 90% RH.

find the total particle light absorption and scattering coefficients. The measurements and calculations presented here are for light extinction by particles, and do not include Rayleigh scattering [Penndorf, 1957] or light absorption by  $\text{NO}_2$  [Hodkinson, 1966].

[15] The single scattering albedo for particles is defined as  $\omega = \sigma_{\text{sp}}/(\sigma_{\text{sp}} + \sigma_{\text{ap}})$ . The results of the previously described calculations are used to determine the single scattering albedo. The particle light extinction properties as a function of relative humidity will be displayed by calculating the particle size distribution and refractive indices as a function of relative humidity and using these values in the Mie calculation.

#### 4. Results

[16] Using the time series of aerosol size and composition measurements made at KCO over the period 11–26 February 1999 by Chowdhury *et al.* [2001], a model for light scattering and absorption was exercised. The results of these calculations then are compared against measured values at KCO. In addition to comparisons between time series of modeled and measured  $\sigma_{\text{sp}}$  and  $\sigma_{\text{ap}}$ , the growth of light scattering with RH, single scattering albedo values, and the

contribution of each chemical species to total light scattering for the externally mixed aerosol case also will be discussed.

[17] Figure 2 shows the time series of light scattering coefficient values at a wavelength of 550 nm due to particles less than 1  $\mu\text{m}$  in diameter. Each panel of that figure provides model results for one RH condition compared to measurements made within a 10% RH band centered at the modeled RH. The model calculation results are shown by boxes. Since the impactor samples were taken over two-day periods, the model value reported for each day of each two day period is the same. The nephelometer data are presented with higher time resolution. Due to the technique used to change RH in the nephelometer, the data at 30 and 90% RH are slightly more sparse than those at 50 and 70% RH. The model and the nephelometer measurements agree very well, with both the model and the measurements showing a peak in the light scattering coefficient for the sampling event spanning 18 February, and a minimum in light scattering during 22 February. The relative error of the model is  $-5.2\%$ ,  $-8.7\%$ ,  $-6.3\%$ , and  $12\%$ , respectively, for the 30, 50, 70 and 90% RH cases. Model results also compare well to measurements at 450 and 700 nm, with slightly lower

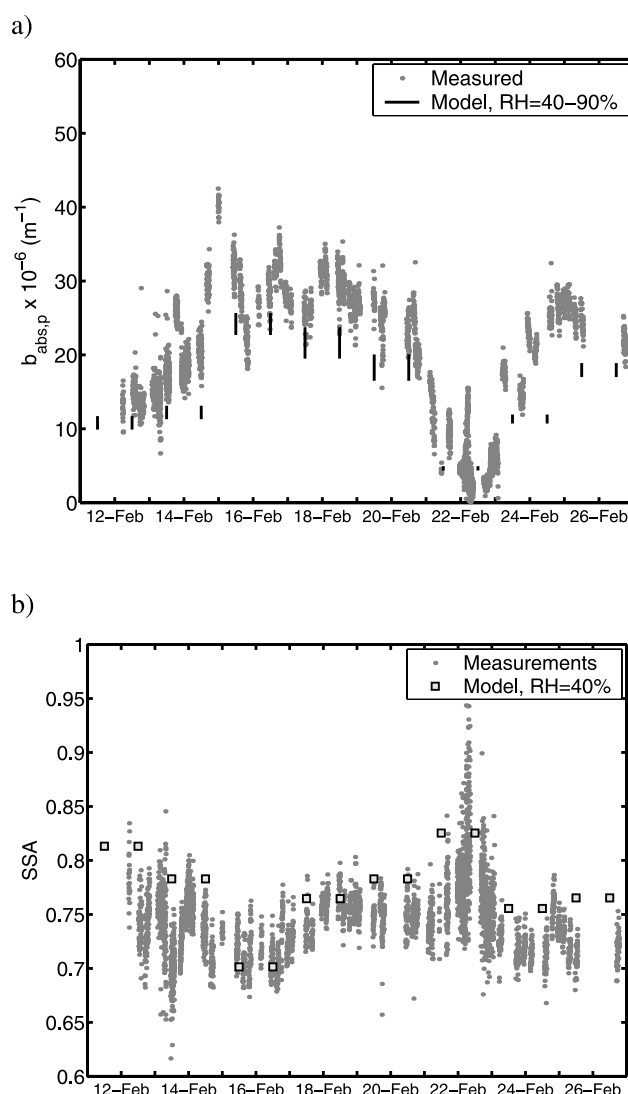


relative errors for the 700 nm case and slightly larger errors for the 450 nm case. Coarse particle data from filter samples are available for the periods of 11–16 February and 24–26 February such that comparisons of modeled and measured values for light scattering by particles smaller than 10  $\mu\text{m}$  aerodynamic diameter can be made at those times. The limited model results are in good agreement with the nephelometer data for particles less than 10  $\mu\text{m}$  in diameter.

[18] An effort was made to understand the impact of uncertainties in the chemical analysis on the light scattering analysis reported here. Chowdhury *et al.* [2001] report the uncertainties on the determination of the concentration of individual chemical species. These have been propagated into uncertainties on the speciation of the samples, and the into uncertainties in the refractive index. Because most of the materials have a very similar real refractive index with the exception of water, the uncertainty in the real refractive index due to uncertainty in speciation is quite small, on the order of 2%. The uncertainty on the real refractive index becomes larger at larger relative humidities when the water is a larger fraction of the mass. When the real refractive indices are reduced by 2%, the calculated extinction coefficients decrease by 5.1 to 12%, depending on the wavelength of light under consideration and the relative humidity. Overall relative humidities and wavelengths studied, the average decrease in the extinction coefficient is 8.2%.

[19] Light absorption coefficient values from the model are compared to measured values in Figure 3a. As was the case for light scattering, the light absorption measurements are made with time resolution on the order of minutes, while the model results are presented for the two day periods over which the impactor samples were taken. The light absorption coefficient measurements in Figure 3a were made near RH of 40% and for purposes of comparison, model calculations are shown over the range of 40–90% RH. Light absorption by particles depends on the imaginary refractive index of the particles which is determined by elemental carbon primarily, with small contributions from soil dust particles. The good agreement between predicted and measured light absorption coefficient values reinforces the value of the elemental carbon data from the impactors. Particle light absorption has only slight RH dependence as shown by the model calculations, with smaller values of the light absorption coefficient predicted for lower relative humidities.

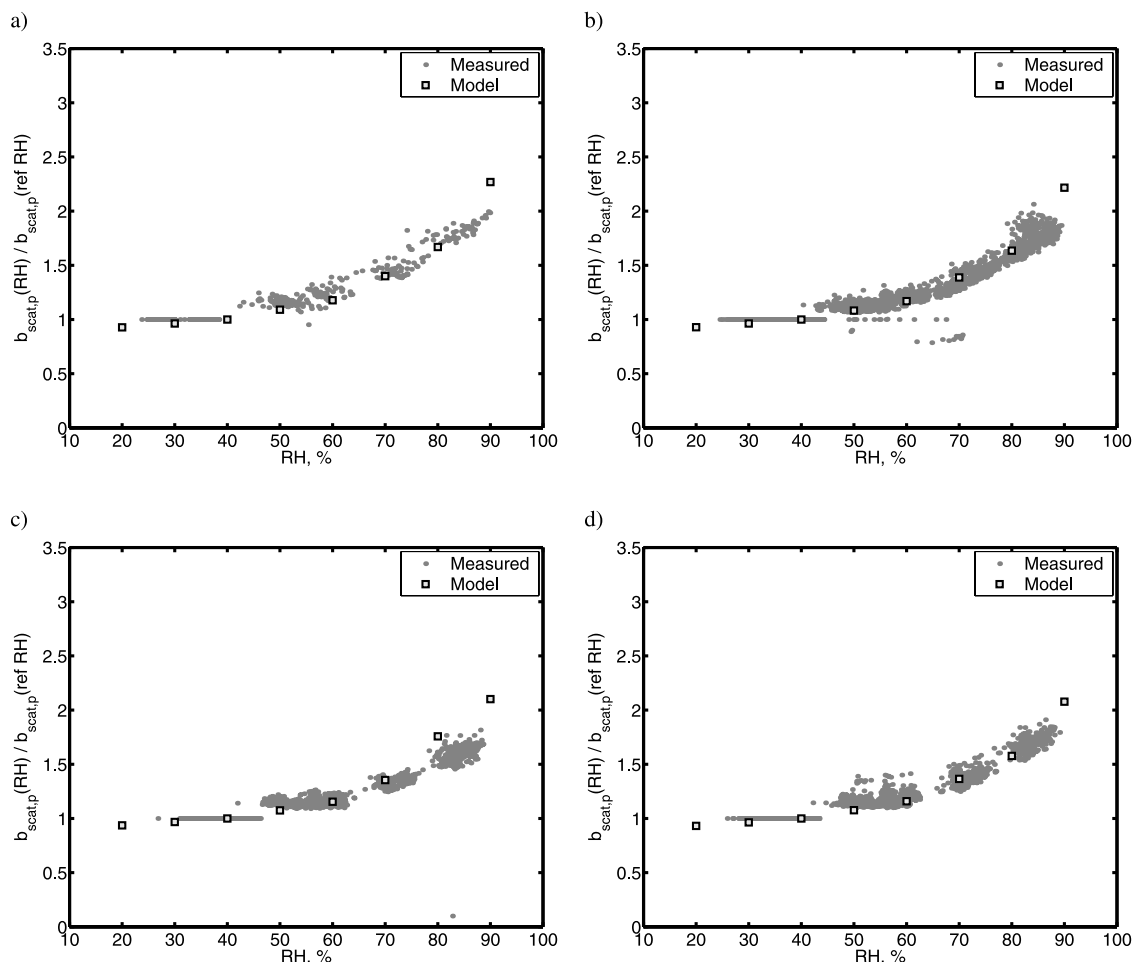
[20] To calculate the single scattering albedo, an important parameter for understanding the potential climate effects of aerosols, light absorption and light scattering measurements at the same RH are required. The light absorption coefficient measurements, which were made at 40% RH, are combined with light scattering coefficient values measured at 40% RH. The measured single scattering albedo ranges from 0.65 to 0.95, with most of the values centered between 0.7 and 0.82. Modeled single scattering albedo values for 40% RH are between 0.68 and 0.81 and the average relative error between the model and the measurements is 4.0%. The single scattering albedo at ambient RH can not be determined from the measurements, but the model, which is capable of predicting the RH dependence of the light scattering and absorption, can be used to predict single scattering albedo values at ambient



**Figure 3.** (a) Light absorption coefficient at 550 nm due to particles less than 1  $\mu\text{m}$  in diameter measured with a PSAP and modeled from the size distributed impactor chemistry. Measurements are made near 40% RH while modeled values for 40 to 90% RH are shown. Panel (b) is the single scatter albedo calculated from light scattering and absorption measurements at 40% RH and modeled values at the same relative humidity.

conditions. Considering two RH values, representative of ambient conditions at Kaashidhoo Island, the range of single scattering albedo values is 0.81 to 0.90 for 80% RH with an average value of 0.86 and ranges between values of 0.86 to 0.93 for 90% RH with an average value of 0.90. These values are similar to the single scattering albedo values of 0.87 to 0.90 reported by Satheesh *et al.* [1999] which were calculated from measurements of light scattering and absorption at KCO in 1998. The modeled single scattering albedo for PM<sub>10</sub> aerosol is a few percent higher than for PM<sub>1</sub>.

[21] In Figure 4 the growth of the light scattering coefficient as a function of RH is presented. The light scattering



**Figure 4.** Relative humidity growth curves for samples collected (a) 11–12 February 1999, (b) 13–14 February, (c) 15–16 February, and (d) 17–18 February. The growth curve is determined from ratio light scattering measurements made with a humidity controlled nephelometer to light scattering at a reference relative humidity of 40%.

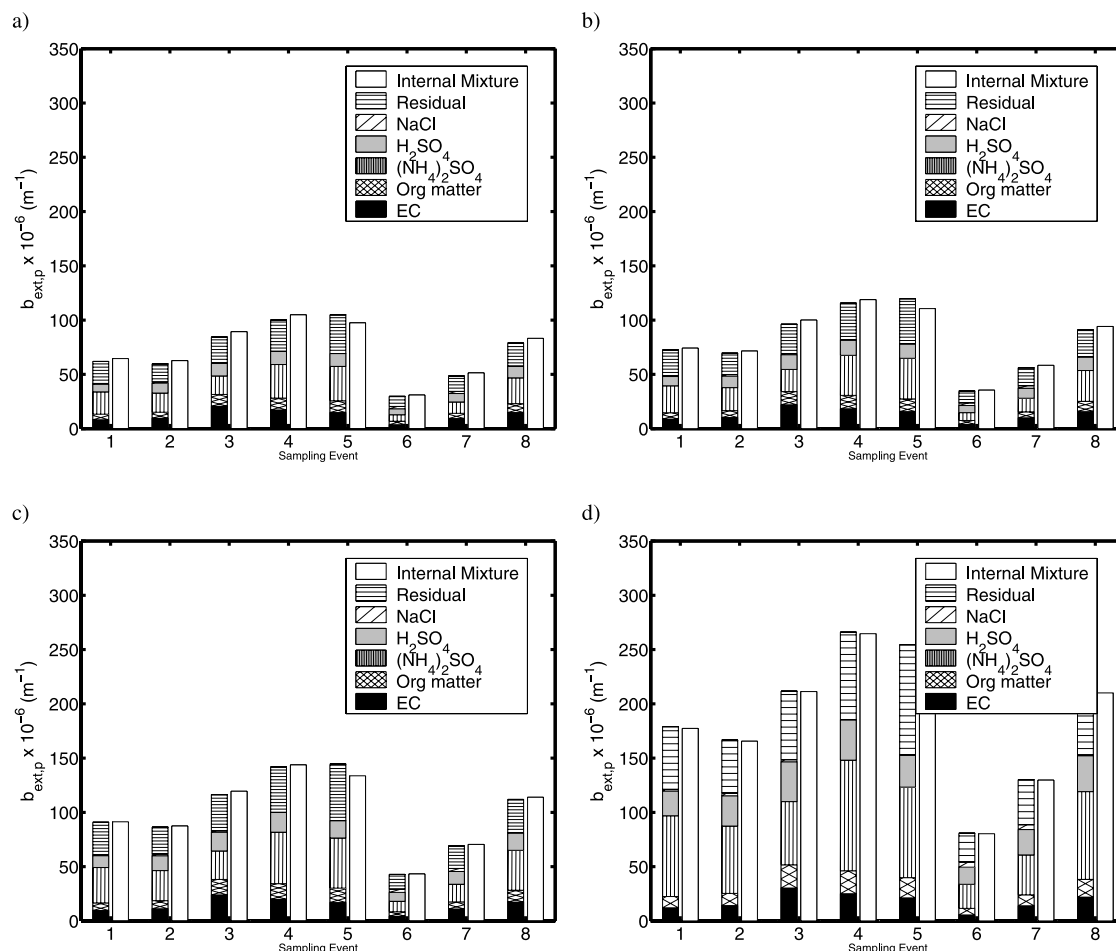
measured by one nephelometer as it is ramped over a range of RH values is normalized by the light scattering coefficient measured with a second nephelometer at a low reference RH. The light scattering coefficient grows larger as water is incorporated into the aerosol as RH is increased, and at 90% RH the light scattering coefficient is about twice that at the low reference RH. Measurements and model results are compared in Figure 4 for the first four sampling events of the campaign, and show that the growth in light scattering and hence particle size with increasing RH is similar for the different sampling events and that this growth can be modeled very well. This is true for all events and for both PM1 and PM10 aerosols. The modeled growth in light scattering is slightly different for each sampling event due to differences between events in the soluble mass of the particles as determined from the impactor chemical composition measurements.

[22] Figure 5 illustrates the results for the particle light extinction calculations using internal and external mixing assumptions. Results are presented for 30, 50, 70, and 90% RH conditions. The modeled light scattering and absorption values are summed to determine the particle light extinction coefficient. The internal and external mixture

models result in very similar predicted values of the particle light extinction coefficient at all RH values for all size fractions. The external mixture model shows that sulfate compounds, organic matter, elemental carbon, and residual (modeled as mineral dust) on average are responsible for 47%, 8%, 12%, and 33%, respectively, of the light extinction by particles at the Maldives Islands modeled at 90% RH. The fraction of light extinction is attributed to the chemical compound and the water associated with that compound. A core and shell model was also used, and compared to the results for the fully internally mixed aerosol model, light scattering decreases by 4–8%, light absorption increases by 3–5%, and overall extinction decreases by a few percent. Compared to the fully internally mixed model, the shell and core model would have slightly worse agreement with the measured light scattering coefficients and slightly improved agreement with measured light absorption coefficients.

## 5. Conclusions

[23] The measured light scattering and absorption in the atmosphere over the Indian Ocean at the Maldives Islands



**Figure 5.** Light extinction coefficients modeled with internal and external mixing assumptions. Results are shown for relative humidities of (a) 30%, (b) 50%, (c) 70%, and (d) 90%.

can be well reproduced with a model for light scattering and light absorption driven by measurements of the airborne particle size distribution and chemical composition. The increase in light scattering at 550 nm as relative humidity increases is reproduced to within  $-9.0$  to  $+12\%$  over the range of relative humidities from 20% to 90%. The measured single scattering albedo for the aerosol is low, averaging 0.86 at 80% RH, and is reproduced by the model with an average relative error of 4%. The model confirms that the large light absorption coefficient values seen at the Maldives Islands during INDOEX are consistent with the measured size distribution and concentration of black elemental carbon particles in the atmosphere at KCO.

[24] The closure calculations performed here demonstrate that light scattering and light absorption at the surface over the Indian Ocean can be calculated directly from particle size and chemical composition data measured at the surface. Air quality models presently exist that can predict particle size and composition from data on source emissions if high resolution emissions data are available [Kleeman and Cass, 1998]. In the future, the effects of changes in source emissions on atmospheric light extinction and hence climate can be calculated if the present model for light scattering and light absorption is combined with a successful model

that can predict aerosol properties based on emissions from sources.

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